

Unconventional ferromagnetic and spin-glass states of the reentrant spin glass $\text{Fe}_{0.7}\text{Al}_{0.3}$

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Spin excitations in a single crystal of $\text{Fe}_{0.7}\text{Al}_{0.3}$ were investigated over a wide range of energy and reciprocal space with inelastic neutron scattering. In the ferromagnetic phase, propagating spin wave modes become paramagnon-like diffusive modes beyond a critical wave vector \mathbf{q}_0 , indicating substantial disorder in the long-range ordered state. In the spin glass phase, the spin dynamics are strongly \mathbf{q} -dependent, suggesting remnant short-range spin correlations.

One class of disordered ferromagnets show reentrant spin-glass behavior [1]: magnetization measurements suggest that the materials change from paramagnetic to ferromagnetic (FM) at the Curie temperature, T_C . Upon further lowering the temperature, the spins are progressively frozen below a freezing temperature, T_f . The low temperature spin frozen state is called a reentrant spin-glass (SG) or mixed state, in which ferromagnetic order is argued to coexist with spin-glass order [2]. For comparison, in a diluted spin-glass, spins freeze directly from a paramagnetic state. Spin waves, the expected collective excitation modes from ferromagnetic long range order, have been studied only at small wave vector, \mathbf{Q} , using inelastic neutron scattering in the FM phase of the reentrant spin-glasses FeCr , AuFe , FeAl , NiMn , $\text{Fe}(\text{Ni,Mn})$ alloys and amorphous $(\text{Fe},T)_{75}\text{P}_{16}\text{B}_5\text{Al}_3$ ($T=\text{Mn, Ni, or Cr}$) [3–7]. The spin waves become broad, decrease in energy, and a quasielastic component appears when the temperature approaches T_f . This short-range quasielastic component continues to increase with lowering temperature in the SG state, as observed in diluted spin-glass such as CuMn [8].

Spin-glasses are generally associated with large degeneracy of the magnetic states caused by disorder, frustration [9] or both. In a reentrant spin-glass, the fact that the FM state occurs at higher temperatures than the SG state suggests a larger entropy for the FM state than for the SG state [1]. This apparent, counter-intuitive situation remains a major mystery in the field of disordered magnetic systems [1]. In this work, using a single crystal, we explore spin dynamics in a greatly expanded \mathbf{Q} and ω range beyond previous neutron scattering studies [10]. We find that the FM state of the reentrant spin-glass $\text{Fe}_{0.7}\text{Al}_{0.3}$ has a qualitatively different spin dynamics behavior from that of a conventional ferromagnetic state. It consists of a mixture of low energy propagating spin waves at small wave vectors and diffusive paramagnon-like spin fluctuations at large wave vectors. This is strikingly reminiscent of the phonon behavior in structural glasses [11]. This confirms that although

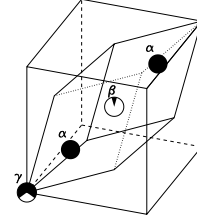


FIG. 1. The fcc lattice structure of $\text{Fe}_{0.7}\text{Al}_{0.3}$. Only atoms inside the primitive unit cell are shown. The α sites are occupied by Fe (black) only. The β site is predominately occupied by Al (white) while the γ site is mostly occupied by Fe.

there is ferromagnetic order, substantial disorders exist at finite length scales in this anomalous ferromagnetic state. In addition, a strongly \mathbf{Q} -dependent spin excitation spectrum exists in the SG state, suggesting remnant short-range spin correlations. Preliminary results were reported at a conference [12].

The single crystal sample of $\text{Fe}_{0.7}\text{Al}_{0.3}$ ($\text{Fe}_{3-\delta}\text{Al}_{1+\delta}$, $\delta = 0.2$) used in this study has a volume of $\sim 1 \text{ cm}^3$ with a mosaic $\sim 1.5^\circ$. It has the face-centered-cubic (fcc) DO_3 structure (space group $Fm\bar{3}m$, No. 225) with four crystallographic sites (refer to Fig. 1): the α sites $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$ and $(\frac{3}{4}, \frac{3}{4}, \frac{3}{4})$ are occupied only by Fe, the β site $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ is occupied predominately by Al with minority Fe ($\sim 9\%$), and the γ site (000) is occupied predominately by Fe ($\sim 70\%$) with minority Al [13]. The lattice parameter is $a = 5.803 \text{ \AA}$ at the room temperature, and there are four formula units in each unit cell. The Curie temperature is $T_C \approx 520 \text{ K}$ and the spin freezing temperature $T_f \approx 80 \text{ K}$ [14].

Neutron scattering experiments were performed at the cold neutron triple-axis spectrometers H9A at the High Flux Beam Reactor (HFBR) of BNL, IN12 at ILL Grenoble, thermal neutron triple-axis spectrometers H7 of HFBR, and BT2 at NIST. Pyrolytic graphite (PG) was used as a monochromator and analyzer. A cold beryl-

lium filter was used for cold neutron measurements and a PG filter was used for thermal neutron measurements to reduce higher order neutrons. Spectrometer configurations used in experiments are specified in the figures. The sample was placed inside an aluminum can filled with He exchange gas in a cryostat. It was aligned with the (hhl) zone coinciding with the scattering plane. We focus in this paper on results from scans along the $[111]$ direction near Bragg points (111) and (000) .

The intensity of the magnetic neutron scattering was measured against a flux monitor placed between the sample and the exit collimator for monochromator. It can be expressed [15] as

$$I(\mathbf{Q}, \omega) = A \cdot f(k_i) \cdot \frac{k_f^3}{\tan \theta_A} \cdot |F(\mathbf{Q})|^2 \cdot \bar{S}(\mathbf{Q}, \omega), \quad (1)$$

where A is approximately a constant for a given spectrometer configuration, k_i and k_f are, respectively, the initial and final wave number for neutrons, $2\theta_A$ is the scattering angle for analyzer, $f(k_i)$ is a correction factor for high order neutrons registered at the flux monitor in a fixed k_f configuration, $|F(\mathbf{Q})|^2$ is atomic form factor for Fe, and $\bar{S}(\mathbf{Q}, \omega)$ is the convolution of the dynamic spin correlation function $S(\mathbf{Q}, \omega)$ with spectrometer resolution function. The polarization factor for \mathbf{Q} along $[111]$ has been absorbed in A . By measuring neutron scattering intensity as a function of momentum transfer $\hbar\mathbf{Q}$ and energy transfer $\hbar\omega$, $\bar{S}(\mathbf{Q}, \omega)$ can be directly determined. For the configurations we used, the four-dimensional convolution yields $\bar{S}(\mathbf{Q}, \omega) \approx S(\mathbf{Q}, \omega)$ for $|\hbar\omega|$ greater than the energy resolution [16]. Factoring out the thermal occupation factor, the imaginary part of the generalized dynamic magnetic susceptibility is given by

$$\chi''(\mathbf{Q}, \omega) = \pi \left(1 - e^{-\hbar\omega/k_B T} \right) S(\mathbf{Q}, \omega). \quad (2)$$

The neutron scattering data shown in this paper are normalized to yield either the $S(\mathbf{Q}, \omega)$ or $\chi''(\mathbf{Q}, \omega)$.

We present first our results at 295 K well within the FM phase. Fig. 2 shows a few examples of constant $\mathbf{Q} = (hhh)$ scans near (000) . The background (refer to the dashed line) is negligible compared to signal. The resolution limited elastic peak at $\hbar\omega = 0$ has been studied before [5] and it will not be discussed here. As observed in previous works near the forward direction [3–7], $S(\mathbf{Q}, \omega)$ peaks at a finite energy. With increasing \mathbf{Q} , the peak energy increases while the damping of the peak becomes stronger. For $h > 0.06$, the spectra are overdamped.

We model the magnetic excitations with a form which is equivalent to the damped harmonic oscillator [17]:

$$\chi''(\mathbf{q}, \omega) = C \left(\frac{\Gamma_{\mathbf{q}}}{\hbar^2(\omega - \omega_{\mathbf{q}})^2 + \Gamma_{\mathbf{q}}^2} - \frac{\Gamma_{\mathbf{q}}}{\hbar^2(\omega + \omega_{\mathbf{q}})^2 + \Gamma_{\mathbf{q}}^2} \right) \quad (3)$$

where C is a constant oscillator strength, and we have chosen $\mathbf{Q} = \boldsymbol{\tau} + \mathbf{q}$, where $\boldsymbol{\tau}$ is the (111) or (000) Bragg

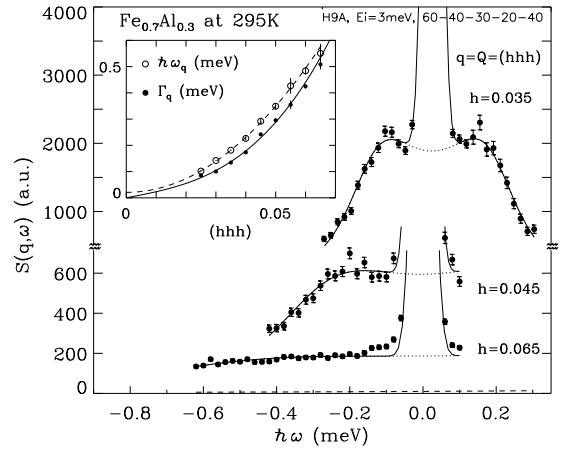


FIG. 2. Small angle inelastic energy scans measured at several \mathbf{Q} values in the FM phase of $\text{Fe}_{0.7}\text{Al}_{0.3}$. Insert: spin wave energy $\hbar\omega_{\mathbf{q}}$ and damping energy $\Gamma_{\mathbf{q}}$ [Eq. (3)] vs $\mathbf{q}=(hhh)$.

point, so that \mathbf{q} is defined within a Brillouin zone. From a standard least-squares fit to each scan, the energy of the spin wave mode, $\hbar\omega_{\mathbf{q}}$, and the damping energy, $\Gamma_{\mathbf{q}}$, are obtained (refer to the insert in Fig. 2). The $\hbar\omega_{\mathbf{q}}$ can be described by

$$\hbar\omega_{\mathbf{q}} = Dq^2 + \Delta \quad (4)$$

with the stiffness constant $D = 37.4(3) \text{ meV}\text{\AA}^2$ and $\Delta = 0.021(2) \text{ meV}$ (refer to the dashed line in the insert in Fig. 2). For comparison, $D = 101 \text{ meV}\text{\AA}^2$ for stoichiometric Fe_3Al . The value for Δ is much smaller than the energy resolution ($\sim 0.07 \text{ meV}$) so it may not be significantly different from $\Delta = 0$. Even in this propagating spin wave regime, the damping energy, $\Gamma_{\mathbf{q}}$, is close to $\hbar\omega_{\mathbf{q}}$ in magnitude.

The range of energy scans is limited by the energy and momentum conservation conditions

$$-\frac{\hbar^2}{2m}(2k_i Q + Q^2) < \hbar\omega < \frac{\hbar^2}{2m}(2k_i Q - Q^2).$$

For \mathbf{Q} larger than $(0.06, 0.06, 0.06)$ (refer to Fig. 2), the accessible energy range can hardly cover $\hbar\omega_{\mathbf{q}}$ or $\Gamma_{\mathbf{q}}$. To obtain reliable measurement of $S(\mathbf{q}, \omega)$, we measure near the Bragg point (111) where a much more extended energy range can be achieved with comparable energy resolution. An example for $\mathbf{q} = (0.08, 0.08, 0.08)$ is shown in the insert in Fig. 3. The sharp peak at -0.52 meV does not appear in a similar scan near (000) , thus it is not part of the magnetic spectrum we are studying. Its origin is still under investigation. The broad magnetic excitations is now clearly overdamped.

For such overdamped magnetic excitations at large q , the value for $\hbar\omega_{\mathbf{q}}$ cannot be obtained reliably. We fixed $\hbar\omega_{\mathbf{q}}$ according to (4) to estimate $\Gamma_{\mathbf{q}}$. The fitting was not very sensitive to $\hbar\omega_{\mathbf{q}}$. Values of $\Gamma_{\mathbf{q}}$ from this and other configurations, spanning more than two orders of magnitude in energy, are shown in the main part of Fig. 3

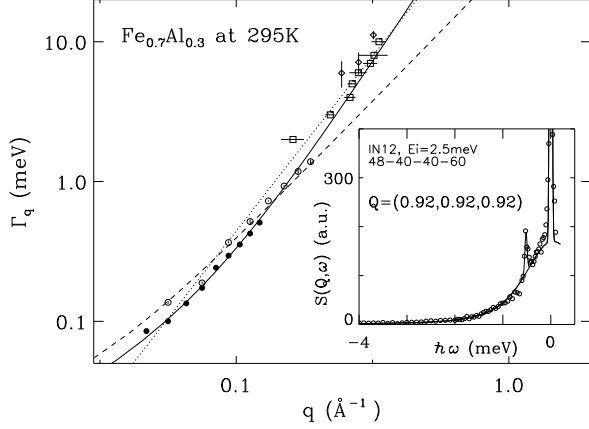


FIG. 3. Damping energy $\Gamma_{\mathbf{q}}$ vs q . The solid line is a fit to Eq. (5) and the dotted line is $\Gamma_{\mathbf{q}} \propto q^{2.5}$. The solid circles are from measurements near (000) at H9A with fixed $E_i = 3$ meV and horizontal collimations 60'-40'-30'-20'-40'; the open circles near (111) at IN12 with $E_i = 2.5$ meV and 48'-40'-40'-60'; the squares near (111) at H7 with $E_f = 14.7$ meV and 40'-20'-40'-40'; and the diamonds near (111) at BT2 with $E_f = 14.7$ meV and 60'-20'-20'-80'. The dashed line is $\hbar\omega_{\mathbf{q}}$ [Eq. (4)] for reference. Insert: a const- \mathbf{Q} scan [$\mathbf{q}=(0.08,0.08,0.08)$] near (111) measured in the FM phase.

using a log-log scale. Roughly, $\Gamma_{\mathbf{q}} \propto q^{2.5}$ (the dotted line). However, the \mathbf{q} dependence of $\Gamma_{\mathbf{q}}$ may be better described by

$$\Gamma_{\mathbf{q}} = \gamma_F q^3 \left[1 + \left(\frac{\kappa}{q} \right)^2 \right] \quad (5)$$

with $\gamma_F = 218(21)$ meV \AA^3 and an inverse length scale $\kappa = 0.073(2)\text{\AA}^{-1}$ (refer to the solid line). This \mathbf{q} dependence for damping is very similar to that for paramagnetic spin fluctuations as observed at $T > T_C$ in ferromagnet MnSi, Pd₂MnSn, Ni and Fe [18]. The damping in the *ferromagnetic* state of Fe_{0.7}Al_{0.3}, thus, appears to share the same characteristics with damping in the thermally disordered *paramagnetic* state of ferromagnets.

To better appreciate this unusual magnetic excitation spectrum, intensity contours of $S(\mathbf{q}, \omega)$ for Fe_{0.7}Al_{0.3} at 295 K, using Eq. (2), (3) and experimentally determined $\omega_{\mathbf{q}}$ and $\Gamma_{\mathbf{q}}$ in (4) and (5), are shown in Fig. 4. There exists a critical wave number, q_0 , where $\hbar\omega_{\mathbf{q}_0} = \Gamma_{\mathbf{q}_0}$. At room temperature,

$$q_0 \approx \frac{D}{2\gamma_F} \left(1 + \sqrt{1 - \left(\frac{2\gamma_F \kappa}{D} \right)^2} \right) = 0.13\text{\AA}^{-1} = 0.07\text{r.l.u.} \quad (6)$$

For $q < q_0$, $\Gamma_{\mathbf{q}} < \hbar\omega_{\mathbf{q}}$. A constant- \mathbf{q} scan yields a peak at a finite energy (c.f., Fig. 2), demonstrating that damped but still propagating spin waves exist. For $q > q_0$, $\Gamma_{\mathbf{q}} > \hbar\omega_{\mathbf{q}}$ and the spin excitation spectrum becomes

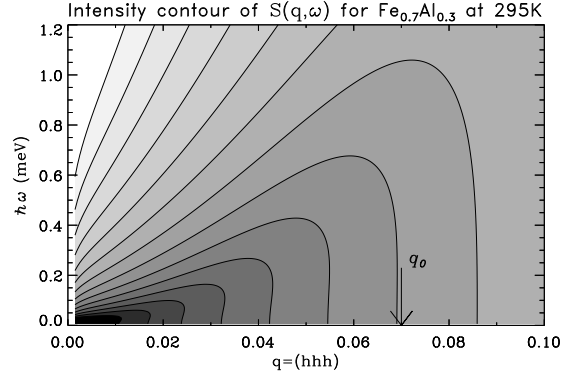


FIG. 4. Intensity contour of $S(\mathbf{q}, \omega)$ in units of $C\text{meV}$ [refer to Eq. (3)] using the experimentally determined $\omega_{\mathbf{q}}$ and $\Gamma_{\mathbf{q}}$ measured at 295 K. Each fading contour level designates a decrease of one order of magnitude. The arrow indicates the critical wave vector.

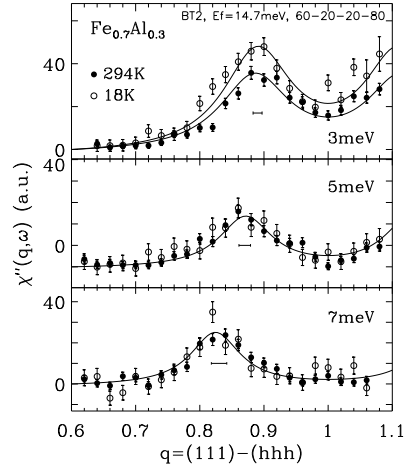


FIG. 5. $\chi''(\mathbf{q}, \omega)$ determined from constant E scans for $\hbar\omega = 3, 5$ and 7 meV. The solid circles were measured at 294 K in the FM phase, and the open circles at 18 K in the SG phase. The horizontal bars indicate the full width at half maximum of the projected instrument resolution function.

overdamped (c.f., insert in Fig. 3). The spectrum in this part of phase space is reminiscent of the paramagnon spectrum of conventional ferromagnetic materials at $T > T_C$ [18]. Specifically, while a constant- \mathbf{q} scan cutting through $S(\mathbf{q}, \omega)$ yields no peak at a finite energy, a constant- $\hbar\omega$ scan shows a peak at finite \mathbf{q} . Some examples of such constant- $\hbar\omega$ scans for Fe_{0.7}Al_{0.3} are shown in Fig. 5. It is interesting to note that a crossover at finite \mathbf{Q} from propagating to diffusive vibrational modes exists in structural glasses [11].

One source of damping is interactions between spin waves [19]. However, at $T = 295$ K $\approx 0.6T_C$, they would not *overdamp* spin waves at a very small wave vector $q \approx (0.07, 0.07, 0.07) = 0.14q_{B.Z.}$, where $q_{B.Z.} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$

is the Brillouin zone boundary. Furthermore, damping due to spin wave interactions approaches zero when $T \rightarrow 0$, while in $\text{Fe}_{0.7}\text{Al}_{0.3}$ damping increases upon cooling [5]. Therefore, spin wave interactions are not the main cause of damping in $\text{Fe}_{0.7}\text{Al}_{0.3}$. Strong damping in this material is most likely due to disorder introduced by random mixture of Fe and Al at the γ and β sites, which at T_f also freezes the spins.

Finally, we compare magnetic excitations at 294 K in the FM phase to those at 18 K in the SG phase. To facilitate such a comparison, data are presented as $\chi''(\mathbf{q}, \omega)$ in Fig. 5 to remove the thermal occupation factor [Eq. (2)]. Spin waves at small \mathbf{Q} near $\tau = (000)$ are known to become overdamped at low temperatures from previous cold neutron studies [3–7]. This fact remains true near the Bragg point (111) [16] and is reflected in the enhanced intensity in the SG state (refer to the open circles) for the $\hbar\omega = 3$ meV scans in Fig. 5. The prevailing view about the diffuse magnetic fluctuations is that frozen spins undergo uncorrelated relaxations. However, unexpectedly, the strong \mathbf{q} -dependent structure of $\chi''(\mathbf{q}, \omega)$ in the FM state remains in the SG state. In fact, the $\chi''(\mathbf{q}, \omega)$ is identical at 18 K and at 295 K for $\hbar\omega \geq 5$ meV. It appears that changes in the dynamic magnetic spectrum in Fig. (4) upon reaching the SG state occur mainly as a result of q_0 decreasing with temperature, so that when T approaches zero, spectral weight at small \mathbf{q} and ω builds up and no propagating features are present. The paramagnon-like spectrum now covers the entire \mathbf{q} - ω space, indicating remnant short-range spin clusters. It is interesting to note that the SG state with frozen disorder exhibits similar spin dynamics as the conventional paramagnetic state where dynamic disorder is caused by thermal energy.

In conclusion, we have characterized over a large ω and \mathbf{q} range the anomalous spin dynamics in the FM phase of the reentrant spin glass material $\text{Fe}_{0.7}\text{Al}_{0.3}$. Like the SG state, the FM state seems also to be a mixed state. In this case, ferromagnetic order and paramagnetic-like disorder coexist at different length scales. In the SG state, the paramagnon-like spin excitations dominate spin dynamics, suggesting short-range spin clusters. This picture, from our experiment, now indicates that the so-called FM state can possess a larger entropy. There likely exists in the phase diagram a crossover between the anomalous ferromagnetic state around 70% Fe and the conventional ferromagnetic state at larger Fe concentrations.

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